



The  
Patent  
Office

Pc/GB 98/00705

3

09/380864

**PRIORITY DOCUMENT**

The Patent Office  
Concept House  
Cardiff Road  
Newport  
South Wales

NP9 TRH	REC'D	20 MAY 1998
WIPO		PCT

I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

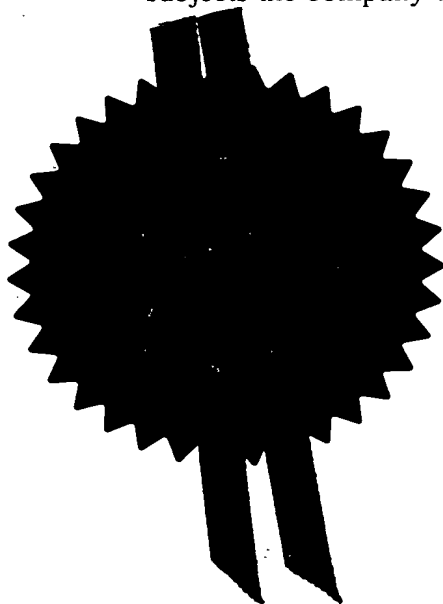
In accordance with the rules, the words "public limited company" may be replaced by p.l.c., plc, P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

Signed

Dated

2 April 1998



**THIS PAGE BLANK (USPTO)**



10-03-97 14:18

01734

P.01

R-173

Job-233

10-MAR-1997 14:17

FROM JOHNSON MATTHEY

TO

901633814444 P.01

Form 1/77

The  
Patent  
Office

1/77

10MAR97 E259275-1 D01091  
P01/7700 25.00

Patents Act 1977  
(Rule 16)

## Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

The Patent Office

Cardiff Road  
Newport  
Gwent NP9 1RH

### 1. Your reference

AA 1335 GB

### 2. Patent application number

(The Patent Office will fill in this part)

9705010.8

10 MAR 1997

### 3. Full name, address and postcode of the or of each applicant (underline all surnames)

JOHNSON MATTHEY PUBLIC LIMITED COMPANY  
78 HATTON GARDEN, LONDON EC1N 8JP, UK

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

536268005

### 4. Title of the invention

IMPROVEMENTS IN EMISSIONS CONTROL SYSTEMS

### 5. Name of your agent (if you have one)

L S BREWER

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

JOHNSON MATTHEY TECHNOLOGY CENTRE  
BLOUNTS COURT, SONNING COMMON,  
READING RG4 9NH

Patents ADP number (if you know it)

560906000

### 6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number  
(if you know it)

Date of filing  
(day / month / year)

### 7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing  
(day / month / year)

### 8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

- a) any applicant named in part 3 is not an inventor, or
  - b) there is an inventor who is not named as an applicant, or
  - c) any named applicant is a corporate body.
- See note (d))

YES

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form. Do not count copies of the same document

Continuation sheets of this form	0
Description	9
Claim(s)	2
Abstract	0
Drawing(s)	0

10. If you are also filing any of the following, state how many against each item.

Priority documents	
Translations of priority documents	
Statement of inventorship and right to grant of a patent (Patents Form 7/77)	
Request for preliminary examination and search (Patents Form 9/77)	1
Request for substantive examination (Patents Form 10/77)	
Any other documents (please specify)	

11. I/We request the grant of a patent on the basis of this application.

Signature

L S BREWER

Date 10 MARCH 1977

12. Name and daytime telephone number of person to contact in the United Kingdom

MRS F E STRANGE 0118 924 2125

**Warning**

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

**Notes**

a) If you need help to fill in this form or you have any questions, please contact the Patent Office on 06 45156666.

b) If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.

d) If you have answered 'Yes' Patents Form 7/77 will need to be filed.

e) Once you have filled in the form you must remember to sign and date it.

f) For details of the fee and ways to pay please contact the Patent Office.

same conditions, a % carbon monoxide conversion of usually greater than 70%, preferably greater than 80%, especially greater than 90%, particularly greater than 95%.

Defining the catalyst systems according to measurement under these conditions does not mean of course that they are necessarily operated under these conditions. In a particular embodiment, the first catalyst system is such that the exhaust gases from the engine flow over it at a low space velocity, particularly below  $40000\text{hr}^{-1}$ . The second catalyst system is usually such that the exhaust gases from the engine flow over it at a space velocity of  $40000\text{-}80000\text{hr}^{-1}$ . The first catalyst system usually contains platinum. The second catalyst system usually contains platinum. Thus, in a particular embodiment each contains platinum. For use, the first catalyst system can be mounted ahead of the second catalyst system in the exhaust apparatus of the engine. The present engine is preferably in a vehicle, for example a passenger car or heavy duty truck.

The skilled person may apply the present invention in a variety of ways.

The first catalyst system may be, for example, a relatively low loading of catalytically active component on a substrate, optionally in combination with components that can retain  $\text{NO}_x$  and/or reducing species, such as zeolite or like absorbents, or alkaline earth metal compounds. We have discovered that reducing the loading of catalytically active component (usually comprising platinum group metal, especially platinum and/or palladium and/or rhodium, particularly platinum, optionally in the presence of base metal components) compared to conventional exhaust gas catalysts, serves to increase the selectivity of the catalyst system towards  $\text{NO}_x$  reduction.

In a particular embodiment, the first catalyst system provides a low space velocity. Normal space velocities for exhaust catalysts systems are 40000-80000hr<sup>-1</sup>. A lower space velocity may be achieved readily in practice by increasing the volume of the catalyst, or by utilising two catalyst "bricks" in parallel.

5

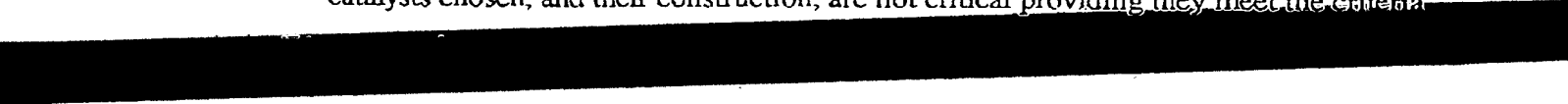
The second catalyst system is required to be of high oxidation activity. Such catalyst need not have NOx selectivity, but must be capable of oxidising hydrocarbons and carbon monoxide under the reaction conditions, to the desired extent, usually so as to satisfy emission standards regulations. Normal space velocities may be used. A suitable oxidation catalyst comprises platinum on a high surface area support, optionally with other components which promote such oxidations.

10

15

20

The skilled person is very familiar with conventional exhaust gas catalyst technology. Generally, a support which is a honeycomb-type extruded ceramic or wound metal monolith or "brick" is coated with a surface area-enlarging washcoat, for example a washcoat consisting of or comprising alumina. Deposited onto the washcoat is a coating of one or more catalytic components, optionally with one or more other components such as ceria, zirconia, zeolite or the like, and the catalyst may be multi-component deposited in discrete layers or some components may be layered, with other components distributed throughout such layers. In the present invention, the actual catalysts chosen, and their construction, are not critical providing they meet the criteria



AA 1335IMPROVEMENTS IN EMISSIONS CONTROL SYSTEMS

5 This invention concerns improvements in emissions control systems, and more especially it concerns improvements in emissions control for engines operating at lean air/fuel ratios.

10 It will be appreciated that with lean-burn engines of various types, including particularly diesel, lean-burn gasoline and direct injection gasoline engines, the control of NO<sub>x</sub> tends to be difficult. This is understandable in that the exhaust gases contain relatively high amounts of oxygen and hence the removal of NO<sub>x</sub> involves reduction of NO<sub>x</sub> to N<sub>2</sub> in an overall oxidising atmosphere. Prior proposals have involved storage of NO<sub>x</sub> in the emission control system until a time when the exhaust gas contains relatively less oxygen, that is until the engine is running "rich", eg during

acceleration. Another proposal is to store unburnt hydrocarbon until a point at which it can be released to contribute to NOx reduction. There remains the need, however, for yet further systems and strategies to achieve control of NOx emissions under lean conditions.

5

The present invention provides a novel emission control system for a lean-burn internal combustion engine, comprising a first catalyst system having relatively high selectivity for NOx reduction, and a second catalyst system having high activity for the oxidation of hydrocarbons and carbon monoxide.

10

The invention also provides a process for the control of emissions from a lean-burn internal combustion engine, comprising passing the exhaust from the engine over a first catalyst system having relatively high selectivity for NOx reduction, then passing the product gases exiting from said first catalyst system over a second catalyst system having high activity for the oxidation of hydrocarbons and carbon monoxide.

15

By selectivity for NOx reduction is meant the ratio of %NOx conversion to % hydrocarbon conversion. The catalyst system having relatively high such selectivity usually has a selectivity of at least 0.2, preferably at least 0.3, especially at least 0.4; this is as measured at a temperature of 230°C, a space velocity of 25000hr<sup>-1</sup> and a hydrocarbon:NOx input ratio of 3:1 counting the hydrocarbon as equivalent propane

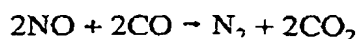
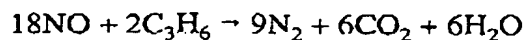
20

for the oxidation of hydrocarbons and carbon monoxide has, as measured under the same conditions, a % hydrocarbon conversion of usually greater than 80%, preferably greater than 90%; it has, as measured under the



It is now well established that carbon monoxide and hydrocarbons play a part in the reduction of NOx. For example, taking the hydrocarbon as C<sub>3</sub>H<sub>6</sub>, the following reactions could take place:

5



10

The molar ratios required for NOx reduction, namely C<sub>3</sub>H<sub>6</sub> to NO and CO to NO, are exceeded over the European test cycle on average with a diesel engine. However, there is competition between NOx and oxygen for the reducing species, and normally only quite low NOx conversions, (eg much less than 10%) are achieved in the European test cycle.

15

It may be preferred to increase NOx conversion under certain conditions by increasing the relative quantity of hydrocarbons in the exhaust. For example, injection of fuel into the exhaust upstream of the first catalyst system may be used. Alternatively, HC storage using zeolites or the like, may be useful. It will be appreciated that there would be a small fuel consumption penalty if fuel injection into the exhaust is used. The high activity second catalyst system is readily capable of catalysing the oxidation of any excess hydrocarbons under the lean conditions.

20

Conventional catalyst manufacturing technology may be used.

The first and second catalyst systems may be mounted in a single "can" in the exhaust system, or they may be separated by a length of exhaust pipe.

The present invention is illustrated by the following Tests.

5

### Test 1

The increase of NOx selectivity corresponding to decreasing platinum loading was shown for a standard 6in (15.2cm) catalyst brick. Exhaust from a 1.9 litre turbo direct injection diesel bench engine, operating at steady state conditions was used. NOx selectivity is measured as % NOx conversion at 230°C/% hydrocarbon conversion at 230°C. The results are shown in Table 1 below.

10

**TABLE 1**

15

Pt loading (g/ft <sup>3</sup> )	NOx selectivity at 230°C
10	1.00
25	0.40
50	0.34
75	0.33
100	0.31

20

A reduced loading of catalyst therefore improves selectivity.

25



**EXAMPLE 1**

A 1996 model passenger car with a 2.5 litre turbo direct injection diesel engine was used with several different exhaust catalyst systems, for standard EUDC emission tests (Extra Urban Driving Cycle emission tests of the European Union). The results are shown in Table 3 below.

**TABLE 3****Catalyst System**

	HC	CO	NOx	HC+ NOx	PM (4)	%NOx
No Catalyst	0.322	1.034	0.394	0.715	0.089	0.0
OEM Catalyst (1)	0.196	0.913	0.389	0.585	0.081	1.3
New Catalyst (2)	0.067	0.336	0.332	0.399	0.079	15.7
Lean-NOx Catalyst (3)	0.054	0.495	0.294	0.347	0.073	25.4
Lean-NOx Catalyst + Oxidation Catalyst (5)	0.037	0.237	0.292	0.329	0.077	25.9

**Notes:**

(1) OEM (Original Equipment Manufacturer) catalyst, 6in long, with 46g/ft<sup>3</sup> Pt.

(2) Advanced oxidation catalyst, 6 in long with 40g/ft<sup>3</sup> Pt.

(3) Lean-NOx catalyst, 12 in long, with 25g/ft<sup>3</sup> Pt.

(4) Particulate matter, g/km

(5) Lean-NOx catalyst, 9in long, with 25g/ft<sup>3</sup> Pt, followed by oxidation catalyst, 3in long, with 100g/ft<sup>3</sup> Pt.

9

All the catalysts used were fresh, *ie* without ageing.

It can readily be seen that the low loading, low space velocity Lean-  
NOx Catalyst is very effective in converting NOx, and that the combination according  
5 to the invention is remarkably effective.

**CLAIMS**

1. An emission control system for a lean-burn internal combustion engine, comprising (A) a first catalyst system, which has a ratio of % NOx conversion to % hydrocarbon conversion of at least 0.2 as measured at a temperature of 230°C, a space velocity of 25000hr<sup>-1</sup> and a hydrocarbon:NOx input ratio of 3:1 counting the hydrocarbon as equivalent propane, and (B) a second catalyst system, which has, as measured under the same conditions, a % hydrocarbon conversion of greater than 80% and a % carbon monoxide conversion of greater than 70%.

2. A control system according to claim 1, wherein the first catalyst system is such that the exhaust gases from the engine flow over it at a space velocity below 40000hr<sup>-1</sup>.

3. A control system according to claim 1 or 2, wherein the second catalyst system is such that the exhaust gases from the engine flow over it at a space velocity of 40000-80000hr<sup>-1</sup>.

4. A control system according to any one of the preceding claims, wherein the first catalyst system and the second catalyst system each contains platinum.

A control system according to any one of the preceding claims, wherein the first catalyst system is ahead of the second catalyst system in the exhaust apparatus of the engine.

6. A process for the control of emissions from a lean-burn internal combustion engine, comprising passing exhaust gases from the engine over a first catalyst system, which has a ratio of % NOx conversion to % hydrocarbon conversion of at least 0.2 as measured at a temperature of 230°C, a space velocity of 25000hr<sup>-1</sup> and a hydrocarbon:NOx input ratio of 3:1 counting the hydrocarbon as equivalent propane, then passing the product gases exiting from the first catalyst system over a second catalyst system, which has, as measured under the same conditions, a % hydrocarbon conversion of greater than 80% and a % carbon monoxide conversion of greater than 70%.

7. A process according to claim 6, wherein the gases are passed over the first catalyst system at a space velocity below 40000hr<sup>-1</sup>.

8. A process according to claim 6 or 7, wherein the gases are passed over the second catalyst system at a space velocity of 40000-80000hr<sup>-1</sup>.

9. A process according to any one of claims 6-8, wherein the first catalyst and the second catalyst system each contains platinum.

10. A process according to any one of claims 6-9, wherein the engine is in a vehicle.

PCT/9B98/00705

JOHNSON MATTHEY

09 MARCH 1998

